8-1

Pergamon

Ann. occup. Hyg., Vol. 41, Supplement 1, pp. 724–727, 1997

Printed in Great Britain
0003-4878/97 $17.00 + 0.00
Inhaled Particles VIII

PII: S0003–4878(96)00105–6

COMPARISON OF PARTICULATE DOSE FROM EXPOSURE TO ENVIRONMENTAL TOBACCO SMOKE (ETS) AND MAINSTREAM CIGARETTE SMOKE USING RADIOTRACERS

J. McAughey, A. Black, J. Strong and C. Dickens
AEA Technology, Aerosol Science Centre, 551 Harwell, Didcot OX11 0RA, U.K.

INTRODUCTION

There has been considerable debate in recent years regarding the health risks resulting from exposure to environmental tobacco smoke (ETS). To date, there has been no direct method of measuring the amount and distribution of tar particulate from ETS deposited in the respiratory tract, or for estimating subsequent clearance. Indeed only one study by Hiller *et al.* (1982) has reported an ETS deposition fraction of 11 ± 4% in a group of five volunteers. It remains difficult to find representative chemical markers of the ETS particulate aerosol in environmental situations, as many have a significant vapour component on ageing and dilution of the smoke.

A ^{123}I-1-iodohexadecane radiolabel for mainstream smoke (Pritchard *et al.*, 1988; McAughey *et al.*, 1996) was unsuitable for sidestream tobacco smoke as 70% of the label (and particulate mass) evaporated on ageing and dilution (Pritchard *et al.*, 1988b).

As an alternative, it is known that the radioactive decay products of the naturally occurring gases ^{222}Rn (radon) and ^{220} Rn (thoron) when first formed have a high mobility and easily become attached to particles such as ETS suspended in the atmosphere. The ^{220} Rn (thoron) series decays via ^{216}Po to ^{212}Pb, with a radioactive half-life of 10.6 h (240 keV γ-ray emission) which would allow monitoring over an appropriate period.

MATERIALS AND METHODS

The methodology for the study of particulate deposition from mainstream cigarette smoke is described in the previous paper (McAughey *et al.*, 1996) with a more detailed description of the radiolabelling method for ^{123}I-1-iodohexadecane and its validation described by Pritchard *et al.* (1988). Details of the ^{212}Pb radiolabel and its validation are described elsewhere (Strong *et al.*, 1994) with the exposure system described in detail by Strong *et al.*, (1994b). In short, the exposure system comprises a ventilated 14 m^3 room, which can be maintained at constant temperature and humidity. Smoke is generated in an external glovebox and fed into the room such that steady-state atmospheres of aged and diluted sidestream smoke at various concentrations can be established and maintained. A ^{228}Th source is also

maintained in a glovebox external to the room to supply ^{220}Rn to a 180 l chamber within the room. The ^{220}Rn rapidly underwent 2 α-decays to produce ^{212}Pb, which attaches principally to the smoke particles present. After a 15 min "growing-in" period, volunteers entered the room and inhaled from the chamber. The inhalation rig used consists of a shuttle valve providing separate pathways for inhalation and exhalation, allowing the collection of exhaled particulate matter; monitoring volume and flow and display of the required inhalation pattern.

Three breathing patterns were used to assess fractional and regional deposition of the labelled surrogate ETS particles; 6 × 1000 ml breaths.min^{-1} mouth breathing and nose breathing and 12 × 500 ml breaths.min^{-1} mouth breathing only.

Nine non-smoking male volunteers were recruited by advertisement, medically examined for fitness to participate in this programme and written informed consent obtained from each. Approval for the study had been granted by the local AEA ethical committee (ATEC) and the Administration of Radioactive Substances Advisory Committee (ARSAC) of the U.K. Department of Health.

Following exposure, the volunteers were monitored over the following 2 days using the collimated whole body monitoring system comprising 6 collimated 15 cm diameter NaI(Tl) detectors described previously (Pritchard and Black, 1988).

RESULTS

The particle size of the ETS aerosol (AMAD) was measured as 0.21 μm (σ_g = 1.32, Delron impactor) and 0.18 μm (σ_g = 1.5, quartz crystal microbalance). Particle concentration was 2.75 × 10^5 particle.ml^{-1} (condensation nucleus counter) and 0.86 mg.m^{-3} (gravimetric), substantially greater than reported environmental levels. However, the main criteria was to achieve high thoron progeny attachment levels to minimise radiation dose to the volunteers.

Total ^{212}Pb deposition in each volunteer was determined by comparing the inhaled with exhaled γ-activity with data shown in Table 1. Whole body measurements showed typical clearance data consisting of two exponential curves. The first curve with half times of approximately 7–10 h represents clearance from the tracheo-bronchial (TB) region of the lung. The second curve with half times of approximately 2 days represents clearance from the pulmonary (P) or alveolar region of the lung. If both curves are projected back to time zero, this gives initial deposition ratios for the respective regions. Regional deposition for the 6 × 1000 ml.min^{-1} mouth breathing pattern are shown in Table 2. Data are compared with values from the LUDEP model (Birchall *et al.*, 1991) based on ICRP 66, 1995.

DISCUSSION

The measured deposition fraction for surrogate ETS differs from that reported by Hiller *et al.* (1982) of 11 ± 4% and values of 80–90% observed for mainstream smoke (U.S. Surgeon General, 1986). However, the Hiller data were for 0.41 μm mass median aerodynamic diameter (MMAD) smoke particles rather than the 0.18–0.21 μm AMAD reported here. Using the LUDEP model (Birchall *et al.*, 1991) to predict total deposition for the experimental conditions reported by Hiller,

Table 1. Total deposition of ETS particulate

Breathing pattern	Deposition (%)	Predicted (%)
6 × 1000 ml.min^{-1}: Mouth	43 ± 17	33
6 × 1000 ml.min^{-1}: Nose	59 ± 10	33
12 × 500 ml.min^{-1}: Mouth	22 ± 8	21

Table 2. Regional deposition (6 × 1 l.min^{-1} mouth)

	Deposition (%)	Predicted
Total	43 ± 16	33
Head (ET)	2.8 ± 1.2	0.6
Thorax	37.2 ± 15	32.4
Bronchial	9 ± 4	6.6
Pulmonary	28 ± 11	25.7
$t_{1/2}$ TB (h)	8.4 ± 2.2	—

a value of 22% was obtained. It can be inferred, from these regional deposition data, that the ETS particulate is depositing in lower generations of the lung than mainstream smoke particulate. This is confirmed by the measured half-time for TB clearance of 8.4 h, which is significantly longer than the values measured by similar techniques in previous studies at Harwell. Clearance values for polystyrene particles (1.5–10 μm diameter) (Pritchard *et al.*, 1986) and for mainstream cigarette smoke (initial particle size 0.7 μm) (McAughey *et al.*, 1996) have all been found with a half-time of approximately 2 h.

The intake of ETS particulate mass remains low. Many authors have used the concept of "cigarette equivalents" in order to give an ETS dose relative to a single cigarette in an active smoker. Estimates of "cigarette equivalents" have varied from 0.024 to 27 cigarette equivalents per day, a range which has been seen as illogical (U.S. Surgeon General, 1986). Calculations for the highest estimate in this report divided the calculated ETS particulate intake by a mainstream smoke intake of 0.55 mg per cigarette, a minimum yield value for cigarette brands available in the U.S. However, the original authors of the cited work quote a sales weighted average mainstream smoke yield of 17 mg per cigarette (Repace and Lowrey, 1980). Use of this more realistic value would reduce the range quoted in the U.S. Surgeon General Report to 0.024–0.9 cigarette equivalents per day. While early estimates based ETS dose on ETS concentration per unit volume multiplied by volume inhaled, more recent calculations of ETS dose have taken into account deposition fraction and adjusted exposure via time-activity matrices, with ventilation volumes adjusted for activity (Holcomb, 1993). These data predict maximum intakes of 109 μg.day^{-1} for men, based on incremental exposure concentrations between smoking and non-smoking areas.

In our calculation exposure data from Holcomb (1993) and Guerin (1992) have been combined with measured data for fractional deposition and ventilation rates. Where total particulate exposure is attributed to ETS, intake in the home or work is estimated at 135 or 187 μg.day^{-1} (or 74 and 61 μg.day^{-1}, respectively for incremental exposure concentrations).

Tar deposition in cigarette smokers using radiotracers have shown an intake of

453 mg per day (range = 307–728 mg per day) in male middle-tar smokers with an average intake of 13.5 mg per cigarette (range = 7.4–22.0 mg per cigarette) (McAughey *et al.*, 1996). This mean delivery of the cigarette was equivalent to the sales weighted average tar for the U.K., consistent with the measured yield of the experimental cigarette. This suggests that daily particulate intake from ETS is of the order of 0.03–0.07% that of cigarette smokers.

However, the regional deposition patterns of mainstream tobacco smoke and ETS particulate are different, with ETS particulate depositing more deeply in the lung. Thus, direct comparisons of particulate retention on a cigarette equivalent basis may be inappropriate. This concern is supported by the U.S. Environmental Protection Agency, who chose not to consider the use of "cigarette equivalents" in their recent publication reviewing the respiratory health effects of passive smoking (USEPA, 1992) for a variety of reasons. For example, although mainstream tobacco smoke (MS) and ETS are qualitatively similar with respect to chemical composition, the absolute and proportional quantities of the smoke components, their physical state and their partitioning between phases can differ. Further differences included variations in particle size between MS and ETS and different breathing patterns in smokers and non-smokers, leading to differences in the distribution and deposition of each type of smoke in the respective populations. Subsequent metabolic differences were also discussed, suggesting dose-response associations were likely to be nonlinear.

CONCLUSIONS

These data are highly significant in terms of risk assessment of ETS particulate exposure, as they clearly demonstrate differences in the deposition pattern in the lung relative to mainstream cigarette smoke. When this is combined with the known physico–chemical differences between the two smoke types, it suggests that extrapolation of exposure and dosimetric data for the two situations is not appropriate.

Acknowledgements—Data from the studies reported were part-funded by the Tobacco Products Research Trust (in respect of mainstream smoking measurements) and the Center for Indoor Air Research (in respect of ETS deposition measurements). The views expressed are solely those of the authors.

REFERENCES

Birchall, A., Bailey, M. R. and James, A. C. (1991) LUDEP: A lung dose evaluation programme. *Rad. Prot. Dosimetry* **38**, 167–174.

Guerin, M. R., Jenkins, R. A. and Tomkins, B. A. (1992) *The Chemistry of Environmental Tobacco Smoke: Composition and Measurement*. Lewis Publishers Inc., Michigan.

Hiller, F. C., McCusker, K. T., Mazumder, M. K., Wilson, J. D., Bone, R. C. (1982) Deposition of sidestream cigarette smoke in the human respiratory tract. *Am. Rev. Respir. Dis.* **125**, 406–408.

Holcomb, L. C. (1993) Indoor air quality and environmental tobacco smoke: concentration and exposure. *Environ. Int.* **19**, 9–40.

ICRP Publication 66 (1995) International commission on radiological protection. *Human Respiratory Tract Model for Radiological Protection*. Pergamon, Oxford.